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Quantum dots: substrate nanopatterning as a path towards the applications

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Abstract

Nanotechnology aims at exploiting the remarkable size effects that arise when materials are reduced to nanoscale dimensions. Exploiting such effects will lead to new applications in different areas of human endeavour. The self assembly of three-dimensional islands is one of the most promising paths towards the fabrication of artificial atoms, or quantum dots (QDs) devoted to nanoelectronic and nanophotonic applications. In order to exploit the unique electronic properties of semiconductor quantum dots in novel quantum effect devices, lateral dimensions of these structures have to be reduced to the order of tens of nm's, the range of De Broglie wavelength of electrons inside these materials. Moreover, millions of quantum dots should be orderly packed in dense arrays to achieve the necessary active volume. So far, the most promising quantum structures have been fabricated using techniques based on self assembling, but their ordering is possible only by appropriate substrate nanopatterning. In this paper we will explore different ways of patterning a substrate and how they affect the growth and ordering of the quantum dots.

1. Introduction

Layer by layer crystal growth by the deposition and self organisation of inorganic material on the surface, called heteroepitaxy, requires the deposited material to reproduce the in–plane lattice features of the substrate material. This generally causes excess strain energy to be stored in the system, if the lattice parameters of the constituents are not matched. The formation of 3D islands, named quantum dots (QD) if their dimension is less than the exciton Bohr radius[1], represents a possible pathway towards a partial strain release, since surface roughening allows for some elastic relaxation.

Fig 1 3D island formation in Ge/Si heteroepitaxy.

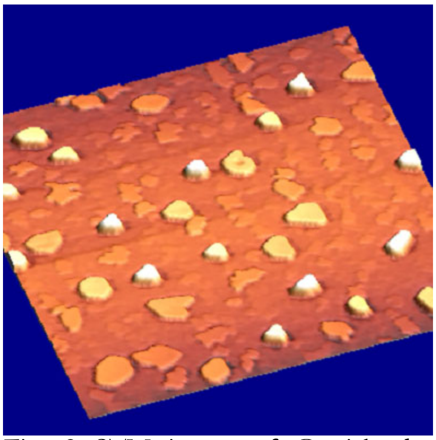

Fig. 2 STM image of Ge islands on Si(111) substrate. Image size: (3x3) m2; vertical scale: 29 nm.

Ge has a lattice constant 4% larger than Si, so that there is a limit up to which the layer-by-layer growth is possible. This limit is usually attained at 3-5 monolayers, depending on the growth temperature, and gives origin to what is usually called "wetting layer". Beyond this limit, the energy of the system can be lowered by the formation of 3D islands (Fig1-2). The growth of this Ge islands on Si substrates is opening the possibility to integrate optoelectronics with today's Silicon technology[2]. Intrinsic silicon is a poor material for optical devices due to its indirect band gap; hence the ability to fabricate optically active elements of Ge (QD) on silicon has great potential for future optoelectronic devices.

There is currently considerable effort being devoted to control size uniformity, density and positioning of such self assembled nanostructures[3,4,5,6]. Different techniques have been developed to achieve nanopatterning without any external processing tool, exploiting the natural strain patterns caused by the heteroepitaxial growth[7,8,9,10], or the step bunching of vicinal surfaces[11].

Based on the analysis of our STM images we report on growth and arrangement of Ge islands on Si(001) substrates nanopatterned using several different approaches.

2. Ordering by self assembly on vicinal surfaces

2.1 Step bunching on Si(111)

On Si(111) surfaces, direct current heating may create bunches of natural surface steps[12], yielding a simple way to obtain a nanopatterned substrate. Several authors have studied this phenomenon. demonstrating that the step configuration at a vicinal surface depends on the direction of the current flowing through the steps, as well as on the miscut angle and on temperature[13,14]. For T > 1220 °C, step bunching (Fig 3) occurs in the step-down direction[15].

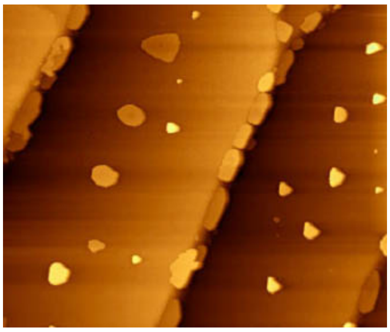

Fig 3 STM image of Ge/Si(111) ordered islands grown by Molecular Beam Epitaxy on self-organized substrate steps at 450 ºC. Image size: 2.9x2.9 μm ^{2}. [Ref. 11]

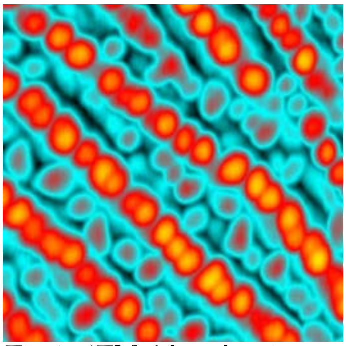

Fig 4 AFM false colour image of nicely ordered ultrasmall Ge islands of lateral size below 50 nm obtained by MBE on a Ge0.5Si 0.5/Si(118) substrate pattern covered by 0.5 ML Sb. Image size 0.5x0.5 μm ^{2} [Ref. 10]

We have analyzed the evolution and distribution of the 3 Dimensional (3 D) islands that form by depositing Ge on this surface at 450 ºC. Initially, triangular islands nucleate and evolve at step edges, rounding their corners up to complete ripening, forming long ribbons. Subsequently, island nucleation takes place at the centre of flat terraces. Ge islands appear to be regularly spaced in scanning tunnelling microscopy images, with an average distance of 360 nm[11].

2.2 Ripples on GeSi/Si(100)

Regular ripples on Si(100) vicinal surfaces can be created by growth instability, exploiting MBE growth of small lattice mismatched GeSi/Si multilayers. It has been recently demonstrated by growing Ge on these surfaces, that periodic ripples, originated by step bunching, macroscopically aligns Ge islands along their direction by providing preferential nucleation sites on their ridges[10]. By using Si(100) substrates 10° misoriented in the [110] direction, and a composition 0.5 of the GexSi1-x alloy we have obtained ripples of 100 nm wavelength10. We have deposited on this substrate a surfactant Sb layer before the final Ge layer. Nice Ge small islands (50 nm typical lateral size) have grown aligned on top of the undulations and at the bottom of the grooves (Fig.4).

3. Nanolithography

In spite of the good results shown in the previous paragraphs, the self-assembling process is still inadequate for the industrial massive production, because of the impossibility to predict the exact nucleation site of the QDs, and of their still large distribution in sizes. Moreover, the efficiency of the devices is largely dependent by the uniformity of the grown structures.

3.1 Holes in the WL by STM

In STM Nanolithography arrays of pits are produced by the STM tip at selected locations, creating preferential nucleation site for islands growth. A real time study of Ge deposition on Si(001) substrates patterned by using the tip of the scanning tunneling microscope has been performed. The experimental observations provide insight into the wetting layer (WL) formation in presence of a regular array of pits. The evolution of a specific hut from WL to pyramid is followed, confirming the existence of a pre-pyramid stage, which evolves with the progressive insertion of {105} facets. Moreover, the results suggest that arrays of intentionally produced pits drive the nucleation process at selected sites. The Si(100) surface was nanopatterned at 500°C by using Scanning Tunneling Microscopy (STM) lithography. At selected positions, with the z-feedback switched-off, pits were elaborated by approaching the STM tip to the surface. The array was re-imaged by the same tungsten tip during the next scan. Pits have diameters ranging from 8 to 15 nm and depths of 1-2 monolayers (ML) and the distance between them is 60±5 nm.

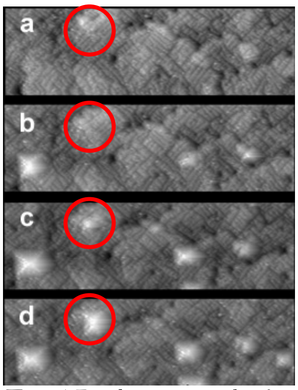

Fig. 5 Real-time growth of pyramids on a nanostructured Si(001) surface. (a)–(g) STM images (250x80x3) nm3 extracted from the movie of Ge deposition at 500°C. [Ref. 29]

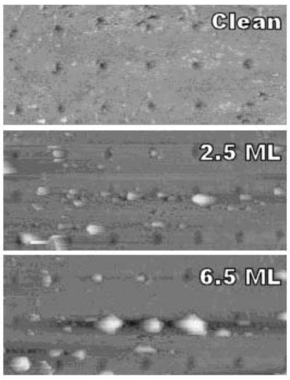

Fig. 6 Sequence of STM images (4.5μmx2.5μm) recorded in real time during Ge deposition on a FIB patterned Si(001) substrate with a 780 nm pitch, starting from the clean surface up to 6.5 ML coverage [Ref.17]

Starting from this kind of surfaces we have followed in real-time the WL growth, the formation of an intermediate stage called pre-pyramid and finally the formation of hut clusters16. From the first stages of deposition during the wetting layer growth it is evident that the Ge atoms do not go inside but on the boundary of the holes.

The 2D-3D transition takes place between 3 and 4 ML of Ge coverage (Fig5). In this sequence, two different stages can be identified: the first stage corresponds to a pre-pyramid (a-b), while the second one to a pyramidal hut (c-d).

3.2 Patterning by Focused Ion Beam

We have studied also Ge growth on Si(001) substrates patterned by Focused Ion beam[17]. Different arrays of holes are produced by FEI Company on the same kind of substrates by using a DualBeam System. The FIB uses a liquid metal ion source to generate a Ga+ ion beam (I ≈1 pA) at a very low distance from the sample surface and applied with energy of 5-25 keV. Two different patterns were created on the Si(001) substrate with a periodicity of 780±30nm (Fig.6). and 500±30nm respectively. After FIB patterning, a chemical cleaning and a thermal annealing are required to reduce Ga contamination below 4x1016cm^{-3}.

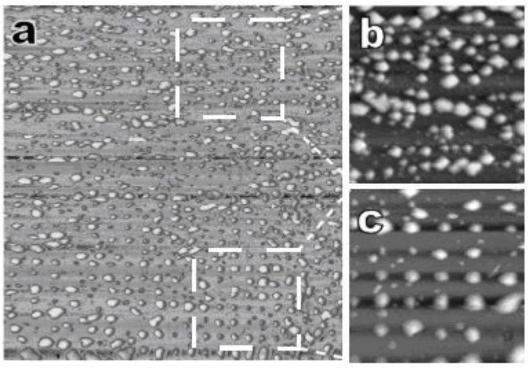

Figure 7: Islands grown on a FIB patterned Si(001) surface after deposition of 20 ML of Ge; (a) STM image (17μmx17μm) showing an unpatterned area (at the top) and a patterned one (at the bottom); zoom (5μmx5μm) (b) on the unpatterned surface and (c) on the FIB patterned surface.

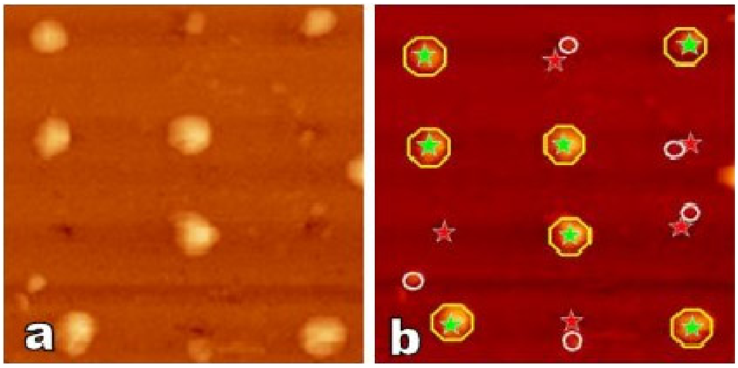

Figure 8: (a) (2.5μmx2.5μm) images of the island morphology and position on a FIB patterned surface showing domes and pyramids. (b) schematics of islands positions of panel (a) showing pyramids as small circles, domes as large octagons and visible/hidden FIB holes as red/green stars.

By following the Ge deposition on a large area we are able to recognize the effect of the patterned array on the Ge growth, comparing patterned and non patterned regions. A large surface area after a Ge deposition of 20ML is imaged in Fig.7 where one can clearly distinguish two domains: an unpatterned area (upper region of Fig 7a) and a patterned area (bottom region of Fig 7a) with a pitch of 780 nm. The difference between the two cases is evident: islands grown on unpatterned region (Fig. 7b) are randomly distributed over the surface. On the contrary in Fig 7c a lateral ordering of islands is observed with a density of 2.4 108 cm^{-2} that is close to the density of pits (2.5 108 cm^{-2}). The Fast Fourier Transform (FFT) made on these two selected regions allows measuring a well-defined periodicity only for Ge islands in the ordered area, equal to 790±50nm. We have also focused our attention on the nucleation site (Fig 8). In Fig. 8b we have schematically shown the corresponding position of pyramids and domes with respect to that of visible and hidden pits. All pyramids (except one) start nucleating nearby a pit and then grow over the pit. We conclude that nucleation starts preferentially at the border of pits. Subsequently each island, increasing its size, evolves to a large dome which covers the underlying pit. At higher Ge coverage, new islands begin to grow in between two domes producing a bimodal distribution[17]. A tighter pattern created on oxidized substrates allows to reach an island density of 4.1 10^{10} cm^{-2}, such is required by nanomemory applications.

Conclusions

We have compared the growth and arrangement of Ge islands on Si(001) substrates nanopatterned using different approaches. The most promising for applications are STM and FIB lithography. The tight pattern created by FIB both on bare and oxidized Si(001) substrates allows to reach island densities required by future applications in highly packed memories.

Acknowledgements

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